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(54) Improved thermoplastic compositions which contain polyolefins and polyphenylene ethers, and articles made therefrom.

(57) An improved polyphenylene ether/polyolefin composition is described, providing excellent properties such as tensile elongation and flame retardance. In addition to the components mentioned above, the compositions also contain an elastomeric block copolymer and a phosphite compound. Another aspect of this invention is an improved electrical conductor article which includes a coating composition of the type described above.

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IMPROVED THERMOPLASTIC COMPOSITIONS WHICH CONTAIN POLYOLEFINS AND POLYPHENYLENE ETHERS AND ARTICLES MADE THEREFROM

This invention relates generally to resinous compositions, and more particularly to thermoplastic blends containing polyphenylene ethers and polyolefins.

Polyphenylene ethers (also known as PPE) are a well-known class of engineering resins which exhibit excellent hydrolytic stability, dimensional stability, toughness, heat resistance, and dielectric properties.

5 In some applications, the overall performance of PPE resins can be enhanced by their admixture with polyolefins, as described, for example, in U.S. Patent 4,166,055, and in the application of G. Lee, Jr. and J. Yates, S.N. 372,218, filed June 23, 1989 and assigned to the assignee of the present application. As an illustration, the use of polyolefins can often improve the impact strength and chemical resistance of PPE-containing compositions.

10 Wire coatings represent one example of a commercial application in which the ductility and dielectric nature of PPE resins is an important attribute. U.S. Patent 4,808,647, for instance, describes curable mixtures which contain PPE and are useful for coating a copper or aluminum wire. The PPE acts in combination with an organic phosphate to enhance the flame retardancy of the coating, in addition to providing some of the other desirable properties mentioned above. Moreover, some PPE-containing
15 compositions may be applied over a base layer of a polymer such as polyvinyl chloride, which has itself already been applied and cured on the metal conductor. The resulting coating structure still meets the physical and electrical requirements for many wire applications.

The increasing interest in wire coatings relates in part to the increasing use of electrical machinery and appliances in homes and businesses. Thus, safety concerns naturally arise, and are often directed to the
20 integrity of the wire coating itself. The end result is a greater need for wire coatings - such as those containing PPE and polyolefin resins - which are much more durable than in the past. For example, they must continue to exhibit a high degree of flame retardance, preferably without using halogen-based agents which sometimes cause corrosion. Furthermore, these coatings must exhibit a high degree of flexibility and "elongation", i.e., the ability to expand and contract without cracking or breaking, even after exposure to
25 high temperatures and/or severe temperature changes. Finally, other properties of the coatings must still be substantially retained, such as toughness and dielectric characteristics.

SUMMARY OF THE INVENTION

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The requirements described above have been satisfied by the compositions of the present invention, which comprise:

- (a) polyphenylene ether resin;
- 35 (b) at least 20% by weight polyolefin resin, based on the weight of the entire composition;
- (c) at least about 1% by weight an elastomeric block copolymer; and
- (d) at least one phosphite compound present in an amount effective to increase the tensile elongation of articles formed from said compositions, by at least about 20% in comparison to that achieved in the absence of the phosphite.

40 As described below, these compositions exhibit attractive characteristics, such as high tensile elongation and good flame retardance.

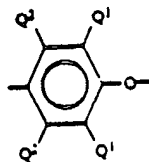
DETAILED DESCRIPTION OF THE INVENTION

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PPE resins are generally well-known in the art, and readily available. Many of them are described in U.S. patents 3,306,874; 3,306,875; and 3,432,469 of Allan Hay; U.S. patents 3,257,357 and 3,257,358 of Gelu Stamatoff; and U.S. patent 4,806,602 of Dwain M. White et al., all incorporated herein by reference.
50 Many PPE resins are also described in two applications for B. Brown et al., serial numbers 210,547 and 210,266, both filed on June 23, 1988 by the assignee of the present invention and incorporated herein by reference. Both homopolymer and copolymer polyphenylene ethers are within the scope of this invention.

The preferred PPE resins are homo- and copolymers which comprise a plurality of structural units of the formula:

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wherein each Q¹ is independently halogen, primary or secondary lower alkyl (i.e., alkyl containing up to 7 carbon atoms), phenyl, haloalkyl, aminoalkyl, hydrocarboxy, or halohydrocarboxy wherein at least two carbon atoms separate the halogen and oxygen atoms; and each Q² is independently hydrogen, halogen, primary or secondary lower alkyl, phenyl, haloalkyl, hydrocarboxy or halohydrocarboxy as defined for Q¹. Examples of suitable primary lower alkyl groups are methyl, ethyl, n-propyl, n-butyl, isobutyl, n-amyl, isoamyl, 2-methylbutyl, n-hexyl, 2,3-dimethylbutyl, 2-, 3- or 4-methylpentyl and the corresponding heptyl groups. Examples of secondary lower alkyl groups are isopropyl, sec-butyl and 3-pentyl. Preferably any alkyl radicals are straight chain rather than branched. Most often, each Q¹ is alkyl or phenyl, especially C₁₋₄ alkyl, and each Q₂ is hydrogen. Suitable polyphenylene ethers are disclosed in a large number of patents.

Especially preferred polyphenylene ethers will be comprised of units derived from 2,6-dimethyl phenol. Also contemplated are PPE copolymers comprised of units derived from 2,6-dimethyl phenol and 2,3,6-trimethyl phenol.

In some embodiments, these compositions may further include vinyl aromatic materials, which are known in the art and described, for example, in U.S. patents 3,383,435; 4,593,058, and 4,661,560, all incorporated herein by reference. These materials are sometimes referred to as polyalkenyl aromatics. Non-limiting examples of the polyalkenyl aromatics are homopolymers such as polystyrene and monochloropolystyrene, as well as modified styrene resins, such as rubber-modified polystyrene (HIPS), the styrene-containing copolymers such as the styrene-acrylonitrile copolymers (SAN), styrene-butadiene copolymers, styrene-acrylonitrile- α -alkyl styrene copolymers, styrene-acrylonitrile-butadiene copolymers (ABS), poly- α -methylstyrene, copolymers of ethylvinylbenzene and divinylbenzene, copolymers of styrene and maleic anhydride, and copolymers of styrene and EPDM (a terpolymer elastomer made from ethylene-propylene diene monomer). Rubber-modified high impact polystyrene is the preferred polyalkenyl aromatic material. A preferred amount of polyalkenyl aromatic material is about 35% to about 55% by weight, based on the weight of the entire composition.

Compositions of this invention further comprise at least one polyolefin resin. Such resins are generally known in the art and are described, for example, in the above-mentioned U.S. Patents 4,166,055, 4,584,334, 4,383,082, in European Patent 0,095,098, in copending application S.N. 372,218, filed June 23, 1989 for G. Lee, Jr. et al., in copending application S.N. 254,519 filed on October 6, 1988 for G. Lee Jr., et al., and on page 836 of The Condensed Chemical Dictionary, Tenth Edition, Van Nostrand Reinhold Company, the entire contents of all of these references being incorporated herein by reference. The polyolefin is usually a polymer formed from an olefin of the general formula C_nH_{2n}. Homo- or copolymers may be used, as well as combinations of polyolefins.

Illustrative polyolefins are polyethylene, poly-propylene, polyisobutylene, copolymers of ethylene and propylene, as well as copolymers of ethylene and organic esters such as ethylene vinyl acetate, ethylene ethyl acrylate, ethylene methyl acrylate, and the like. The preparation of these polyolefins is generally well-known in the art.

The preferred polyolefins are polyethylene, polypropylene and polybutylene. Especially preferred are low density polyethylene (LDPE), linear low density polyethylene (LLDPE), medium density polyethylene (MDPE), high density polyethylene (HDPE), and high impact (rubber-modified) polypropylene.

The amount of polyolefin present in the composition should be at least about 20% by weight, based on the weight of the entire composition. In preferred embodiments, the level should be at least about 30% by weight, and in the most preferred embodiments, should be at least about 40% by weight.

Another component of this invention is an elastomeric block copolymer. The block copolymers are generally well-known in the art and described, for example, in U.S. Patent 4,080,356 of Gergen et al., U.S. patents 4,166,055 and 4,584,334, and in European Patent 0,095,098, all incorporated herein by reference. Examples of commercially available block copolymers are the Kraton® series of polymers, available from Shell Oil Company.

Preferred block copolymers for the present invention are characterized by an A-B, A-B-A', or (AB)_m-X structure, or mixtures of these structures, wherein A and A' are each polymerized vinyl aromatic hydrocarbon blocks, each B is a hydrogenated, partially hydrogenated, or non-hydrogenated block derived from at least one polymerized conjugated diene, X is the radical of a multifunctional coupling agent, and m is an integer of at least 2. The block polymers preferably contain between about 15 and 50% by weight of vinyl aromatics.

In some preferred embodiments, A and A' are independently selected from the group consisting of styrene, alpha-methyl styrene, para-methyl styrene, vinyl toluene, vinyl xylene, and vinyl naphthalene; and B is selected from the group consisting of butadiene, isoprene, ethylene, butylene, 1,3-pentadiene, 2,3-dimethyl butadiene, and combinations thereof.

Each block A and A' usually has a molecular weight in the range of about 2000 to 100,000, while each block B usually has a molecular weight in the range of about 25,000 to 1,000,000.

An especially preferred elastomeric block copolymer for this invention comprises blocks of styrene and ethylene-butylene, and is often referred to as "SEBS".

The preparation of elastomeric block copolymers is known in the art and can be effected, for example, by successive polymerization of the monomers in solution in the presence of a monolithium hydrocarbon initiator.

Furthermore, the olefinic double bonds of the block copolymers can be selectively hydrogenated in any conventional manner, e.g., the use of molecular hydrogen and catalysts based on metals or salts of metals of group VIII of the Periodic Table. Further aspects of the preparation of the elastomeric block copolymers can be found in European Patent 0,095,098, in U.S. Patents 3,113,986 and 3,700,633, in German Public Disclosure 1,222,260, and in German Letters of Disclosure 2,013,263.

The elastomeric block copolymer is used in an amount which enhances some of the properties of these compositions. This amount is usually at least about 1% by weight, based on the weight of the entire composition. More preferably, the block copolymer level is at least about 5% by weight, and most preferably, at least about 15% by weight. The exact amount will of course depend on several factors, such as the particular composition of the compatibilizer and the type of polyolefin being used.

The phosphite compound used in the present invention can be of various types, such as alkyl phosphites, aryl phosphites, alkyl-aryl phosphites, diphosphites, polyphosphites, and thiophosphites. Many are disclosed in U.S. Patents 4,659,760 and 4,472,546, incorporated herein by reference. Examples of suitable phosphites are triphenyl phosphite, diphenyl phosphite, trimesityl phosphite, dimesityl-phenyl phosphite, trineopentyl phosphite, didecylphenyl phosphite, dichloroethyl phosphite, tributyl phosphite, trilauryl phosphite, tris(nonylphenyl) phosphite, tridecyl phosphite, diphenyldecyl phosphite, tricresyl phosphite, triisooctyl phosphite, tri-2-ethylhexyl phosphite, 1 trioctadecyl phosphite, thiophosphite, phenyl-diethyl phosphite, phenyl-di-(2-ethylhexyl) phosphite, isooctyldiphenyl phosphite, diisooctylmonophenyl phosphite, distearyl pentaerythritol diphosphite, octadecyl 3-(3,5-di-t-butyl-4 hydroxyphenyl) propionate (eg., ULTRANOX® 276 additive), bis(2,4-di-t-butylphenyl) pentaerythritol diphosphite (commercially available as ULTRANOX® 626 or ULTRANOX® 624), diphenyl isooctyl phosphite, diisooctyl phosphite, distearyl phosphite, diphenylisodecyl phosphite, diisodecyl pentaerythritol diphosphite, poly(dipropylene glycol) phenyl phosphite, and trilauryl trithiophosphite, as well as mixtures of the above.

The phosphite compound is present in an amount effective to substantially increase the tensile elongation of materials made from these compositions. A "substantial" increase is defined here as an increase in tensile elongation of at least about 20%, as determined according to UL 1581, compared to the materials without the phosphite. A part of this invention is the discovery that relatively small amounts of the phosphite compound are required for unusually high increases in tensile elongation in the particular compositions described herein. Furthermore, these compositions surprisingly maintain the high tensile elongation values after being heat-aged.

The effective amount of phosphite compound is usually at least about 0.01% by weight, based on the weight of the entire composition. A more preferred amount for many requirements, especially when the composition of this invention is used as a wire coating, is at least about 0.1%. Usually, the phosphite is used in a quantity no greater than about 10%. The most appropriate range can be determined according to various factors, such as the contemplated end use for the product, as well as the particular identity of the other components in the composition.

In some embodiments, a plasticizer is included in the composition. Suitable plasticizers include phosphate compounds, mineral oil, low-melting solids such as phthalate, adipate, and sebacate esters; polyols such as ethylene glycol, and the like.

The aromatic phosphates are the preferred plasticizers. Examples include triphenyl phosphate, isopropylated triphenyl phosphate, and isobutylated triphenyl phosphate, as well as mixtures of these

materials. The aromatic phosphates are usually used at up to about 25% by weight, based on the weight of the entire composition. A preferred level is about 1% by weight to about 15% by weight.

A wide variety of other additives may be included in the compositions of this invention, their utility being of course dependent in part on the contemplated end use for the compositions. Non-limiting examples of these additives include flame retardants, reinforcing agents such as fibrous glass and graphite whiskers, antioxidants, mold release agents, mineral fillers, pigments, dyes, abrasion resistant compounds, and the like. These additives are usually included in amounts of between about 1% and 50% by weight of the total composition.

These compositions can be prepared by well-known procedures. Often, the ingredients are combined as a pre-mix blend, and then extruded on a single or twin screw extruder.

Another aspect of this invention is an improved electrical conductor article, which includes a coating material comprising the components set forth above, i.e., PPE, polyolefin, elastomeric block copolymer, and phosphite compound, applied over the surface of an electrical conductor, for example, copper or aluminum wire or cable. (Sometimes, the conductor is first coated with a material which inhibits or prevents oxidation). The coating composition can be applied by known methods, such as extrusion, followed by cooling. The coating thus forms an excellent electric-insulator for the metal. If desirable, one or more outer or "jacket" layers may be formed over the coating. These layers are also often formed of a polymeric material, such as polyvinyl chloride, a polyester like polybutylene terephthalate, or a polyamide, and can also be applied by extrusion techniques, such as tandem extrusion.

Alternatively, the coating composition of this invention may be used itself as a jacket layer applied over a wire insulating material such as polyvinyl chloride.

It should be understood that the composition of this invention is not limited for use as a coating material. It may be used to form any type of molded article where excellent tensile elongation and flame retardance are desirable.

The following examples are provided to illustrate various embodiments of this invention. It is to be understood, however, that the embodiments are given for the purpose of illustration only, and do not and should not be regarded as limiting the invention to any of the specific materials or conditions described therein.

EXAMPLES

The compositions set forth in Table 1 were prepared by dry-blending the ingredients and then melt-blending the mix, using a single screw extruder. All values are by weight, unless otherwise indicated.

The extrudate was quenched and pelletized, and then remelted in an extruder equipped with a die for coating wires. The molten polymer was thereby applied to a copper wire.

Tensile tests were performed on the material after it was stripped from the wire. Some of the material was heat-aged for 7 days at 136°C and then cooled at room temperature for about 24 hours before being tested for tensile characteristics.

As mentioned above, the tensile values set forth below were determined by UL 1581.

Samples 2,3,5 and 6 are part of the present invention; while samples 1 and 4 are controls.

TABLE 1

5						Tensile Values ^(g)				
						Initial ^(f)	After Heat Aging ^(h)			
	Sample No.	PPE	Phosphite ^(b)	Elastomeric ^(c) Block Copolymer	Polyolefin ^(d)	Phosphate ^(e)	T.S.	T.E.	T.S.	T.E.
10	1*	30 ^(a)	0.0	20	50	10	3.1	158	4.3	176
	2	30	0.5	20	50	10	3.7	242	4.8	216
	3	30	0.5	20	50	10	4.0	243	5.5	228
	4*	30	0.5	20	50	10	—	145	—	144
15	5	30	0.5	20	50	10	—	204	—	195
	6	30	0.5	20	50	10	—	197	—	215

*controls

(a) All values in parts by weight

(b) Samples 2, 5: Tridecyl Phosphite

20 Samples 3, 6: Bis(2,4-di-t-butylphenyl) pentaerythritol diphosphite

(c) Styrene-Ethylene-butylene-Styrene block Copolymer

(d) Linear Low Density Polyethylene

(e) Triphenyl Phosphate

25 (f) "TS" = Tensile Strength; "TE" = Tensile Elongation

(g) Tensile Values Determined According to UL 1581

(h) Heat Aging for 7 Days at 136°C

30 The data in Table 1 demonstrate that the addition of phosphites to the compositions resulted in a very large increase in tensile elongation. Furthermore, the increase was generally maintained after heat aging of the samples.

Modifications and variations of the present invention are possible in light of the above teachings. It is understood, however, that such changes are within the intended scope of this invention, as defined by the
35 appended claims.

Claims

40 1. A thermoplastic composition comprising:

a) polyphenylene ether resin;

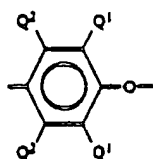
b) at least about 20% by weight polyolefin resin, based on the weight of the entire composition;

c) at least about 1% by weight of an elastomeric block copolymer, based on the weight of the entire composition; and

45 d) a phosphite compound present in an amount effective to increase the tensile elongation of articles formed from said compositions, by at least about 20% in comparison to that achieved in the absence of the phosphite.

2. The composition of claim 1, wherein the polyphenylene ether comprises a plurality of structural units of the formula:

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and in each of said units independently, each Q¹ is independently halogen, primary or secondary lower alkyl, phenyl, haloalkyl, aminoalkyl, hydrocarbonoxy, or halohydrocarbonoxy wherein at least two carbon atoms separate the halogen and oxygen atoms; and each Q² is independently hydrogen, halogen, primary or secondary lower alkyl, phenyl, haloalkyl, hydrocarbonoxy or halohydrocarbonoxy as defined for Q¹.

- 5 3. The composition of claim 2 wherein each Q¹ is an alkyl group having from 1 to 4 carbon atoms, and each Q² is hydrogen.
4. The composition of claim 2 wherein the polyphenylene ether is derived from the group consisting of 2,6-xylenol, 2,3,6-trimethylphenol, and mixtures of these materials.
5. The composition of claim 2 wherein the polyphenylene ether is poly(2,6-dimethyl-1,4-phenylene ether).
- 10 6. The composition of claim 1 wherein the polyolefin resin is selected from the group consisting of low density polyethylene, linear low density polyethylene, medium density polyethylene, high density polyethylene, polypropylene, rubber-modified polypropylene, ethylene vinyl acetate, ethylene ethyl acrylate, ethylene methyl acrylate, and mixtures of any of the foregoing.
7. The composition of claim 1 wherein the polyolefin level is at least about 30% by weight, based on the weight of the entire composition.
- 15 8. The composition of claim 7 wherein the polyolefin level is at least about 40% by weight, based on the weight of the entire composition.
9. The composition of claim 1 wherein the elastomeric block copolymer is present in an amount of at least about 5% by weight, based on the weight of the entire composition.
- 20 10. The composition of claim 1 wherein the elastomeric __block copolymer is characterized by an A-B, A-B-A', or (A-B)_m-X structure, or mixtures of these structures, and A and A' are each polymerized vinyl aromatic hydrocarbon blocks, each B is a hydrogenated or non-hydrogenated block derived from a polymerized conjugated diene, X is the radical of a multifunctional coupling agent, and m is an integer of at least about 2.
- 25 11. The composition of claim 10 wherein A and A' are independently selected from the group consisting of styrene, alpha-methyl styrene, para-methyl styrene, vinyl toluene, vinyl xylene, and vinyl naphthalene; and B is selected from the group consisting of butadiene, isoprene, 1,3-pentadiene, and 2,3-dimethyl butadiene.
12. The composition of claim 10 wherein the elastomeric block copolymer comprises blocks of styrene and ethylene-butylene.
- 30 13. The composition of claim 1 wherein the phosphite compound is present in an amount of at least about 0.01% by weight, based on the weight of the entire composition.
14. The composition of claim 13 wherein the phosphite level is at least about 0.1% by weight, based on the weight of the entire composition.
15. The composition of claim 13 wherein the amount of phosphite compound is in the range of about 0.01% to about 10% by weight, based on the weight of the entire composition.
- 35 16. The composition of claim 1 wherein the phosphite compound is selected from the group consisting of alkyl phosphites, aryl phosphites, alkyl-aryl phosphites, diphosphites, polyphosphites, thiophosphites, and mixtures of any of the foregoing.
17. The composition of claim 16 wherein the phosphite compound is selected from the group consisting of triphenyl phosphite, tridecyl phosphite, triisodecyl phosphite, diphenyl phosphite, diisooctyl phosphite, distearyl phosphite, bis(2,4-di-t-butylphenyl) pentaerythritol diphosphite, distearyl pentaerythritol diphosphite, and mixtures of any of the foregoing.
- 40 18. The composition of claim 1, further comprising a vinyl aromatic polymer.
19. The composition of claim 1, further comprising a plasticizer.
- 45 20. The composition of claim 19 wherein the plasticizer is selected from the group consisting of phosphate compounds, mineral oil, polyols, phthalate esters, adipate esters, sebacate esters, and mixtures of any of the foregoing.
21. The composition of claim 20 wherein the plasticizer is selected from the group consisting of triphenyl phosphate, isopropylated triphenyl phosphate, isobutylated triphenyl phosphate, and mixtures of any of the foregoing.
- 50 22. The composition of claim 1, further comprising a reinforcing agent.
23. The composition of claim 22 wherein the reinforcing agent is glass.
24. An improved electrically conductive article, comprising:
 - a) an electrical conductor;
 - 55 b) a coating disposed over the conductor, and formed from a composition which comprises:
 - i) polyphenylene ether resin;
 - ii) at least about 20% by weight polyolefin resin, based on the weight of the entire composition;
 - iii) at least about 1% by weight of an elastomeric block copolymer, based on the weight of the entire

composition; and

iv) a phosphite compound present in an amount effective to increase the tensile elongation of the coating, by at least about 20% in comparison to that achieved in the absence of the phosphite.

25. The article of claim 24, wherein the coating is applied directly to the conductor surface.

5 26. The article of claim 25, wherein an outer layer of a polymeric material is disposed on the surface of the coating.

27. The article of claim 24, wherein the coating is applied over an insulating layer of a polymeric material which itself is applied directly on the conductor surface.

28. The article of claim 24, wherein the composition further comprises a material selected from the group
10 consisting of phosphate compounds, mineral oil, polyols, phthalate esters, adipate esters, sebacate esters, and mixtures of any of the foregoing.

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EUROPEAN SEARCH REPORT

Application Number

EP 90 11 4041

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
Y	EP-A-0 243 761 (BORG-WARNER) * page 1, line 33 - page 4, line 17; claims 1-13 * - - -	1-28	C 08 L 71/04
D,Y	US-A-4 383 082 (LEE,JR) * column 1, line 51 - column 3, line 55; claims 1-7 * - - -	1-28	
D,Y	EP-A-0 095 098 (BASF) * page 3, line 14 - page 11, line 5; claim 1 * - - -	1-28	
A	US-A-4 293 660 (SUGIO ET AL) * column 1, line 51 - column 4, line 7; claims 1-7 * - - -	1-12	
A	EP-A-0 326 895 (GENERAL ELECTRIC) * page 2, line 38 - page 5, line 7; claims 1-9 * - - -	1-12	
D,A	US-A-4 584 334 (LEE,JR. ET AL) * column 1, line 48 - column 3, line 33 * - - -	1	
A	US-A-4 255 321 (BRUSSEN) * column 2, line 15 - column 3, line 44; claims 2, 3 * - - -	1,13	
A	US-A-3 449 292 (SNEDEKER) * column 1, line 55 - column 6, line 60; claims 1-13 * - - - - -	1,13	
The present search report has been drawn up for all claims			
Place of search		Date of completion of search	Examiner
The Hague		31 January 91	BOURGONJE A.F.
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone		E : earlier patent document, but published on, or after the filing date	
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